Journal of Cleaner Production 222 (2019) 760–767

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Three considerations for modeling natural gas system methane emissions in life cycle assessment

Emily A. Grubert [a, *], Adam R. Brandt [b]

[a] Civil and Environmental Engineering, Georgia Institute of Technology, United States
[b] Energy Resources Engineering, Stanford University, United States

ARTICLE INFO

Article history:
Received 27 August 2018
Received in revised form
7 March 2019
Accepted 8 March 2019
Available online 13 March 2019

Keywords:
Methane
Inventory
Climate change
Carbon footprint
Leakage
Natural gas

ABSTRACT

Natural gas is a fossil fuel accounting for about 30% of US primary energy consumption. Climate change is one of the primary environmental issues associated with natural gas use: natural gas combustion releases carbon dioxide. A less emphasized issue is that natural gas is mostly methane, a potent greenhouse gas (GHG). The climate impact of natural gas use is thus sensitive to the amount of methane that escapes from the natural gas system unburned. We call attention to three considerations for modeling natural gas-related methane emissions in life cycle assessment (LCA). First, natural gas system methane leakage is inconsistently characterized and likely systematically underestimated by commonly used life cycle inventory (LCI) databases. Second, studies are often imprecise in assumptions about process boundaries. This matters because not all natural gas uses rely on the same infrastructure and induce the same methane leakage. Third, there is not yet a stable estimate for the global warming potential (GWP) of methane. Newer estimates tend to be larger, which further exacerbates the underestimation of GHG impacts from natural gas systems. Data uncertainty is common in LCA, but natural gas-related methane emissions deserve special attention due to their influence on a decision-relevant parameter (GHG intensity) in product systems across the economy.

© 2019 Elsevier Ltd. All rights reserved.

1. Introduction

Natural gas accounts for about 30% of US primary energy consumption (EIA, 2018) and is widely used for electricity generation, heating, and industrial purposes. Given its prevalence, natural gas is part of the life cycle of a large number of product systems. Natural gas is primarily comprised of methane (CH_4), the second most significant GHG for anthropogenic climate change, after carbon dioxide (CO_2) (Weyant et al., 2006). Methane has a relatively high global warming potential (GWP): even a small amount of emitted methane can result in substantial carbon dioxide-equivalent (CO_2e) GHG emissions, one of the most commonly studied environmental indicators (Grubert, 2017). Questions about the amount of methane that escapes to the atmosphere unburned from natural gas systems, which we will call methane leakage, are thus highly relevant to environmental impact evaluations like life cycle assessment (LCA).

Despite recent studies on methane leakage from natural gas systems, most of which suggest that leakage is underestimated (Alvarez et al., 2018; Balcombe et al., 2018; Brandt et al., 2014; Farquharson et al., 2017; Heath et al., 2014; Littlefield et al., 2016; Sanchez and Mays, 2015; Zhang et al., 2014), little attention has been paid to the impact of methane leakage inventories on product carbon footprints. Natural gas is an indirect input to many products via electricity, heating, and chemical feedstock supply. As these inputs are typically background processes for product-specific LCAs, database defaults are often used. This ubiquity means that inaccurate methane leakage inventories are widely relevant to LCA and, especially given the small number of processes involved, should be priorities for LCA database improvement.

1.1. Methane leakage from natural gas systems

We will use the terms methane “leakage” or methane “emissions” interchangeably to refer to all methane emitted unburned from natural gas systems. Not all leakage is unintentional: for example, some safety systems are designed to release gas when a system becomes overpressured due to unusual operating conditions or equipment problems. Leakage is generally a combination of non-purposeful emissions (sometimes called fugitives) and

* Corresponding author. 790 Atlantic Dr NW, Atlanta, GA, United States.
E-mail address: gruberte@gatech.edu (E.A. Grubert).

https://doi.org/10.1016/j.jclepro.2019.03.096
0959-6526/© 2019 Elsevier Ltd. All rights reserved.

designed or purposeful emissions (sometimes called vents).

Researchers have been concerned for decades about methane leakage from natural gas systems and its influence on climate change (Lelieveld et al., 2005, 1993; Meier et al., 2005), with substantial disagreement about the value and dynamics of leakage rates (Kirchgessner et al., 1997). Data from outside North America are sparse (though see Yacovitch et al., 2018, a recent study from the Netherlands), neglecting possibly significant regional variability (Bouman et al., 2015; Gibon et al., 2017). For example, a study comparing GHG emissions from coal versus shale gas-fired power generation in China uses United States (US) emissions factors due to a lack of Chinese data (Chang et al., 2015). Recent literature has suggested that methane emissions might be systematically underestimated by official inventories, in part because a substantial portion of emissions is due to large, infrequent, and unintentional releases that can be difficult to detect (Brandt et al., 2016, 2014). The exact volume of leakage is not precisely understood (Alvarez et al., 2012; Brandt et al., 2014; Burnham et al., 2012; Hausfather, 2015, 2014; Heath et al., 2014; Jeong et al., 2014; Lelieveld et al., 2005, 1993) due to regional variability (Allen et al., 2013; Jiang et al., 2011; Karion et al., 2015, 2013; Pétron et al., 2014), operational practices, regulatory context, and other factors.

The energy analysis community has primarily focused on leakage while examining whether substituting natural gas for coal in the electricity sector will reduce overall climate change impacts (Farquharson et al., 2017; Gilbert and Sovacool, 2017; Hausfather, 2015; Howarth et al., 2011; Roy et al., 2015). Less attention has been paid to the relative GHG impacts of natural gas-versus electricity-based residential and commercial applications like heating and cooking. Although leakage from the natural gas distribution system has been studied (Costello, 2014; Hendrick et al., 2016; Jackson et al., 2014; McKain et al., 2015; Phillips et al., 2013), end use leakage from equipment, homes and businesses is rarely assessed (for an exception see Lavoie et al., 2017). This lack of study is particularly apparent for residential applications where methane leakage from appliances might be environmentally relevant in a comparison across heating options (Oliphant, 1994).

1.2. Three claims about methane leakage in LCA databases

In this paper we make three claims with respect to the treatment of methane in LCA databases.

First, commonly used inventory databases are inconsistent and likely systematically underestimate methane leakage from the natural gas system. Major ambiguities within and across inventory databases are common. We examine how existing databases model methane leakage from the natural gas system and quantify the relevance of natural gas system leakage rates to the GHG footprint of basic materials to demonstrate the scope of this problem. Our results show that methane leakage from natural gas systems should be a priority target for inventory improvements.

Second, databases (and individual studies) have unclear or incorrect assumptions about which natural gas system processes are applicable for a given supply chain. This is important, as it affects both how users might respond to leakage and what methane emissions are assigned to a given use of natural gas.

Third, there is not yet a stable estimate for the GWP of methane, affecting comparability of LCA studies over time. Newer estimates of methane's GWP tend to be larger. Analysts studying comparative energy systems should perform sensitivity analysis on the GWP and should report actual methane masses rather than CO_2e only, to allow forward compatibility and comparability of results.

The analysis below examines and supports these claims in order. We will focus on US methane leakage in LCA databases, though many of the same issues apply to carbon footprinting or environmental impact assessment studies.

2. Methods

2.1. Methane leakage in life cycle inventory databases

Given the vast amount of data needed for LCA and a desire for consistency across studies, LCA practitioners often use one of a few common life cycle inventory (LCI) databases. For major databases, we ask: 1) What do the databases assume about US natural gas system leakage rates? And 2), how significant is embodied natural gas-related methane leakage for GHG footprints of non-natural gas product systems?

2.1.1. Database assumptions

We examine leakage assumptions in several common LCI databases. Our primary goal is to illustrate current status and discrepancies rather than to provide a set of recommended values. We do, however, incorporate synthetic insights from the many recent studies that have estimated leakage outside the context of life cycle inventories (Abrahams et al., 2015; Allen et al., 2014, 2013; Alvarez et al., 2018; Balcombe et al., 2018; Brandt et al., 2014; Ge et al., 2016; Heath et al., 2014; Hendrick et al., 2016; Jeong et al., 2014; Jiang et al., 2011; Karion et al., 2015, 2013; Lavoie et al., 2017; Littlefield et al., 2016; Mitchell et al., 2015; Pétron et al., 2014; Phillips et al., 2013; Zimmerle et al., 2015).

We examine three databases, each of which is the most current version available:

1) Economic Input-Output Life Cycle Assessment (EIO-LCA) Model (Carnegie Mellon University Green Design Institute, 2008), 2002 Producer Price model;
2) The National Renewable Energy Laboratory (NREL) United States Life Cycle Inventory database (USLCI) (NREL, 2012); and
3) The ecoinvent database, version 3.5 (ecoinvent, 2018).

USLCI and EIO-LCA data are older and less frequently updated than ecoinvent data, but relevantly, these databases are free while ecoinvent is not. Thus, it is reasonable to expect that they remain in use. Some other common LCA tools base their US natural gas-related methane emissions on databases we assess, so these are implicitly assessed as well. For example, GREET uses EPA inventories and GaBi/Thinkstep uses the USLCI as source data.

In each case, we: 1) identify unit process data for US-based natural gas systems; 2) match available inventory processes with processes in the US natural gas system to evaluate coverage; 3) convert data from each database's inventory format to a mass of methane basis, using process-specific data on energy densities, pressure, and standard gas conditions; and 4) calculate methane emissions as a mass percentage of gross methane withdrawals for each included process.

To investigate the accuracy of LCI data, we compare them with three non-LCI leakage estimates: 1) a recent inventory for 2015 natural gas supply chain methane emissions based on facility-scale ground measurements and aircraft observations (Alvarez et al., 2018); 2) Environmental Protection Agency (EPA) greenhouse gas inventory (GHGI) estimates for 2013 (EPA, 2015); and 3) EPA GHGI estimates for 2015 (EPA, 2017). EPA GHGI mass emissions are converted to leakage rates using the method of Brandt et al. (2014). Year 2015 EPA GHGI results are the basis for many leakage parameters in Argonne National Laboratory's GREET model (Burnham, 2017), and these data reflect a recent change in calculation methodology for natural gas systems. EPA 2013 GHG data are also included because they are of a similar vintage to the USLCI (released in 2012) dataset.

2.1.2. Significance beyond natural gas systems

We are interested in LCI data on methane leakage from the natural gas system largely because of the hypothesis that these background data have a large influence on GHG footprint results beyond the natural gas system itself. To test this hypothesis, we investigate the contribution of natural gas system methane leakage to the GHG footprints of six illustrative materials: plastic, fertilizer, aluminum, steel, electricity, and cement. These materials were chosen because they are themselves common inputs for products throughout the economy. Demonstrating that natural gas system methane leakage matters for the GHG footprints of these materials thus indicates its relevance for GHG footprinting more generally.

2.2. The location of methane leakage

Not all processes that are part of the natural gas supply chain apply to all uses of natural gas. For example, leakage from the natural gas distribution system (a low-pressure network delivering natural gas to typically nonindustrial end users, like homes) does not in most cases affect the carbon intensity of natural gas-fired electricity from power plants. Most natural gas-fired power plants are supplied from the high-pressure transmission system either directly or via high pressure laterals (personal communication, Dynegy Investor Relations, March 2018). We illustrate the relevance of this system structure using natural gas consumption data from the Energy Information Administration (EIA) to provide guidelines on how natural gas end uses map to natural gas system subprocesses (EIA, 2017).

2.3. The influence of global warming potential on methane leakage impact

Use of different GWPs in the literature means that methane emissions reported on a CO_2e basis, even evaluated over the same time horizon, are not necessarily comparable. To illustrate the impact of this instability, we examine methane's GWP in the five Intergovernmental Panel on Climate Change (IPCC) Assessment Reports (the First, Second, Third, Fourth, and Fifth are referred to as FAR, SAR, TAR, AR4, and AR5) (Intergovernmental Panel on Climate Change, 2018). We do not address the separate question of which time horizon to use when comparing climate impacts of methane to carbon dioxide (Farquharson et al., 2017; Gilbert and Sovacool, 2017; Hausfather, 2015; Howarth et al., 2011; Roy et al., 2015).

3. Results and discussion

3.1. Methane leakage in life cycle inventories

3.1.1. Database assumptions

Table 1 summarizes the leakage assumptions in studied LCI databases. The supplementary material (SM) contains calculation details.

For each life cycle stage, leakage is reported as a cumulative mass percentage of gross withdrawals of the methane that reaches each stage. That is, a value of 1.4% under "distribution" means that for natural gas in the distribution system, life cycle methane emissions to that point are 1.4% of the mass of the gross withdrawals of methane that reached the distribution system, not that 1.4% of methane is lost at the distribution stage or that 1.4% of gross methane withdrawals have been lost due to distribution and upstream processes. Percent emissions are computed as follows (Equation (1)):

$$\text{percent emissions through stage } n = \sum_{i=1}^{n} \frac{\text{mass methane emitted at stage } i}{\text{mass methane reaching stage } i \times \frac{\text{mass methane withdrawn (incl. losses)}}{\text{unit mass methane reaching stage } i}} \quad (1)$$

Leakage rate estimates for a stage can be computed by subtracting the cumulative losses listed for upstream processes. Note also that gross withdrawals of methane are not equal to gross withdrawals of natural gas because natural gas is not pure methane. Relatedly, the embodied noncombustion GHG intensity of natural gas is also affected by the presence of other GHGs in withdrawn natural gas, including CO_2 and ethane. These nonmethane contributions are outside the scope of this study but could be relevant for some systems, particularly those using natural gas sources containing large mole fractions of CO_2.

Table 1 indicates several issues with the LCI databases. First, all investigated databases likely underestimate methane emissions from natural gas systems: total leakage reported by each database is similar to EPA estimates, which evidence suggests are too low (e.g., Alvarez et al., 2018). Also, they disagree. For example, data from ecoinvent (dated 2018) are much different from USLCI data, even though documentation for ecoinvent suggests its values are based on the USLCI. USLCI and EIO-LCA data are both based on US federal data from the late 1990s and early 2000s, but leakage rates by process are meaningfully different.

Second, databases have variable coverage of the natural gas system. This matters in part because LCA is sometimes used for hotspot identification, allowing analysts to focus on life cycle stages with the most potential for improvement. For example, even though the EIO-LCA production leakage of 0.57% is not that different from the USLCI production + processing leakage of 0.54%, users would draw very different conclusions about where to focus leakage reduction efforts. Note that none of the investigated databases account for methane leakage associated with nonindustrial end users (for example, residential cooking and heating), though its existence has long been understood (Oliphant, 1994) and can affect the climate implications of appliance choices.

Third, databases include numerous documentation discrepancies and nonintuitive definitions. Minor changes in interpretation or approach result in very different values for Table 1. For example, although ecoinvent 3.5 shows a leakage rate slightly higher than EPA estimates, this value is highly sensitive to a nonobvious parameter related to natural gas production from gas versus oil wells: through ecoinvent 3.3, use of a different value for that parameter, but essentially identical data otherwise, led to implied leakage of 1.2% rather than 1.6%. Even in version 3.5, the parameter does not reflect actual US conditions (see SM, S1.4). In another example, in EIO-LCA, calculating emissions based on the "Pipeline transportation" process suggests transmission-stage leakage of 0.42% (when emissions are allocated to coproducts based on monetary value) to 1.6% (when emissions are allocated to coproducts based on the 2002 EPA Greenhouse Gas Inventory), not the 0.34% implied by decomposing the "Natural gas distribution" sector (see SM, S1.2).

Table 1
Methane leakage as cumulative mass percentage of gross withdrawals of methane reaching a given stage, assuming natural gas progresses through each stage in sequence.

Data source	Production	Processing	Transmission & Storage	Distribution	End Use
Reference estimates					
Alvarez et al. (2018)	2.0%	2.1%	2.4%	2.5%[a]	n/a[b]
EPA (2015)	0.82%	0.93%	1.1%	1.3%	n/a
EPA 2013	0.38%	0.63%	1.0%	1.4%	n/a
LCA databases					
EIO-LCA[c]	0.57%	n/a	0.91%	1.4%	n/a
USLCI	0.34%[d]	0.54%	0.92%	n/a	n/a
ecoinvent v3.5	1.3%[e]	n/a	1.6%	n/a	n/a

[a] Note Alvarez et al., 2018 estimates for distribution leakage are based on the 2017 EPA GHGI, which they consider a lower bound for the true value (see Alvarez et al., 2018, S1.5).
[b] "n/a" denotes a process that is not explicitly included and where no data are available.
[c] Methane leakage from the natural gas system is based on the furthest downstream sector "Natural gas distribution:" see SM, S1.2 for details.
[d] The 0.34% value cited is based on the process "natural gas, extracted," which documentation suggests includes all natural gas production. The process "natural gas, at extraction site" is associated with 1.4% leakage. The distinction between the two is unclear: documentation indicates that the database distinguishes between associated and nonassociated natural gas, but both "natural gas, at extraction site" and "natural gas, extracted" indicate in their "technology description" that the process includes both natural gas from natural gas-only wells (nonassociated) and from oil wells (associated). See SM, S1.3 for details.
[e] Based on life cycle inventory data. Note that the production value is based on the mass-weighted percentage of contributions from "natural gas production" (methane leakage: 1.8%; ecoinvent contribution: 70%) and the natural gas allocation of "petroleum and gas production, on-shore" (methane leakage: 0.1%; ecoinvent contribution, 30%). See SM, S1.4 for details.

In USLCI and ecoinvent, ambiguous process names and definitions can easily lead to unintended choices with large implications for results. In both cases, there are multiple processes for natural gas extraction with unclear or no guidance on which to use. In USLCI, natural gas produced from gas-only wells versus from any well appear to be differentiated by the terms "extracted" (0.34% leakage) and "at extraction site" (1.4% leakage), respectively. Process descriptions are nearly identical, and both imply that all natural gas (both associated and nonassociated) is included. Further, "natural gas, extracted" uses a mass-denominated output while "natural gas, at extraction site" uses a volume-denominated output, so users might choose one or the other based on the units they are using without realizing the data are different. This introduces unnecessary possible confusion.

A similar issue exists for ecoinvent users, who might notice that the cumulative value for leakage post-transmission is smaller than leakage associated with "natural gas, production," even though the reference flow for the two is the same: 1 m^3 of "natural gas, high pressure." This discrepancy exists because the post-transmission process is also drawing on a second production process, "petroleum and gas production, on-shore" (see SM, S1.4). The use of the term "natural gas, high pressure" to refer to different systems at different places in the inventory is confusing. As with USLCI, users likely will not anticipate that data associated with "natural gas, production" are very different from those associated with the natural gas output of "petroleum and gas production, on-shore:" life cycle methane leakage for these processes is 1.8% or 0.10%, respectively. Less ambiguous names would help (e.g., calling one flow "natural gas, high pressure, at oil and gas co-production facility" and the other "natural gas, high pressure, at gas production facility").

Another serious source of confusion is use of the term "natural gas" itself. Without data on the composition of natural gas, converting among mass, energy, and volume units can introduce serious errors. Further, users might reasonably assume that the type of "natural gas" being referenced is relevant to the process at

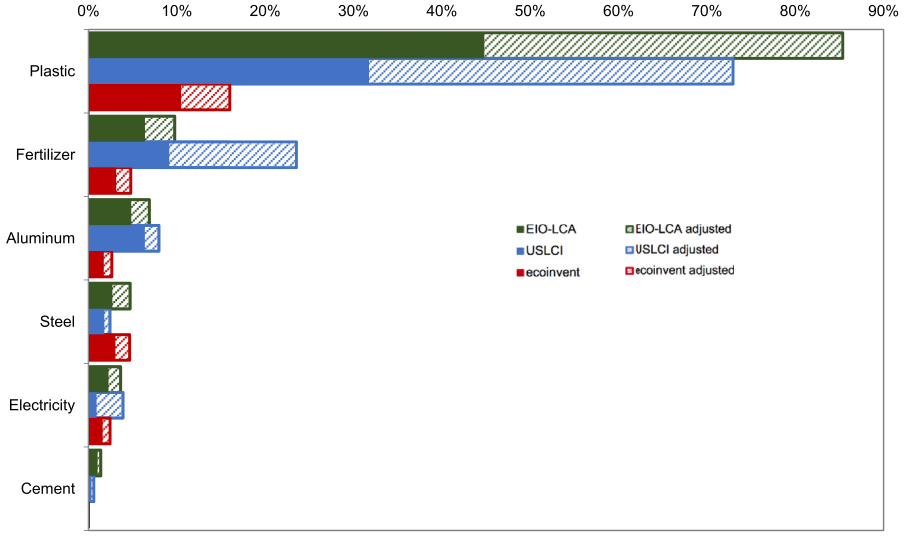

Fig. 1. Contribution of natural gas system methane leakage to greenhouse gas intensity for six basic materials in three life cycle inventory databases.
Notes: Specific processes used to proxy the six materials are found in the SM Data File. "Plastic" is "Plastics material and resin manufacturing" for EIO-LCA and polyethylene terephthalate (PET) for USLCI and ecoinvent. "Fertilizer" is "Fertilizer manufacturing" for EIO-LCA, "nitrogen fertilizer" for USLCI, and "urea ammonium nitrate" for ecoinvent. "Electricity" is the broadest category identified for US electricity in each case. "Adjusted" values show recalculated values assuming that leakage rates in each database matched values from Alvarez et al., 2018.

hand, which is not always the case. For example, in ecoinvent 3.5, the stated mass density of "natural gas, high pressure" of 0.84 kg/m^3 implies that the reference flow is raw gas at standard temperature and pressure, not pipeline-quality gas at high pressure (see SM, S1.4). USLCI uses energy rather than mass densities to describe flows but has similar problems (see SM, S1.3). Additionally, databases rarely document whether energy densities are reported on a higher heating value (HHV, sometimes called "gross") or lower heating value (LHV, sometimes called "net") basis. The difference between these two measures is about 10%, and the impact might not be noticed outside the energy community (e.g., one should not expect that a polymer chemist performing an LCA of a novel plastics process would know to check for this definitional detail).

3.1.2. Significance beyond natural gas systems

Problems with the characterization of methane leakage from natural gas systems are important for GHG footprinting beyond studies focused on natural gas itself. To illustrate this point, we use the EIO-LCA, USLCI, and ecoinvent databases to estimate natural gas system methane leakage GHG emissions as a percentage of direct process GHG emissions for six major industrial materials: plastic, fertilizer, aluminum, steel, electricity, and cement (Fig. 1). Calculation details, including the exact processes referenced, can be found in the SM Data File. Fig. 1 uses a GWP of 34 for methane (AR5, 100 year, with climate carbon feedback) and also presents "adjusted" values that show what results would be using leakage rates from a recent empirical study of US natural gas system leakage (Alvarez et al., 2018).

For some materials, embodied methane emissions from natural gas systems are not large compared to direct emissions. For example, cement manufacturing releases substantial process CO_2 emissions and uses little natural gas, so the influence of leaking methane is negligible. For others, however, most notably plastic, embodied methane leakage is substantial.

Fig. 1 suggests that correctly representing natural gas-system methane leakage should be a priority for LCI databases. Only a small number of processes need be updated, a minor task relative to the potential benefits to accuracy. Ensuring these emissions are visible in LCA studies can also help make results actionable. Reducing methane leakage from natural gas systems is likely an easier mitigation opportunity (Gallagher et al., 2015; Gladd, 2016; Hopkins et al., 2016; Lamb et al., 2015; Ravikumar and Brandt, 2017; von Fischer et al., 2017) than reducing byproduct emissions inherent to the production of a product (e.g., from calcining or

			Life Cycle Stages Relevant to End Use					
End Use	**Description[1]**	**2016 Consump-tion, Bcf**	**Produc-tion**	**Process-ing**	**Trans-mission**	**Storage**	**Distri-bution**	**User leakage[2]**
Lease fuel	natural gas used for production	1,169	yes					
Plant fuel	natural gas used for processing	421	yes	yes				
Pipeline and distribution use[3]	natural gas used for transmission and distribution pipelines	697	yes	yes	some	some	some	
Transmission	*not reported separately*	*n/a*	*yes*	*yes*	*some*	*some*		
Distribution	*not reported separately*	*n/a*	*yes*	*yes*	*yes*	*yes*	*some*	
Residential	natural gas used in residences	4,345	yes	yes	yes	yes	yes	yes
Commercial	natural gas used in commercial settings	3,105	yes	yes	yes	yes	yes	yes
Industrial	natural gas used in non-power generation industrial settings	7,722	yes	yes	yes	yes	*rare*	yes
Vehicle Fuel	natural gas used in vehicles	41	yes	yes	yes	yes	yes	yes
Electric Power	natural gas used for electricity generation	9,983	yes	yes	yes	yes	*rare*	yes
Approximate end use volume affected, %[4]		27,486	~100%	~96%	<90%	<90%	<23%	~87%

Fig. 2. Relationship between end uses of natural gas in the United States and upstream processes for which leakage and other impacts are embedded. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Notes: Bcf = billion cubic feet. Green/yes = "upstream process for this end use," yellow/some = "upstream process for some of this end use," where "some" means that neither condition is unusual, red = "not an upstream process for this end use," where "rare" means that analysts should assume the upstream process does not apply unless they specifically know about an unusual situation. Source: https://www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_a.htm.

[1]Formal definitions at https://www.eia.gov/dnav/ng/TblDefs/ng_cons_sum_tbldef2.asp.

[2]All users have the potential to emit fugitive (leaked) methane. These emissions vary widely by customer type and by specific customer: averages should be used very carefully, with effort to match the leakage rate to the end use type. Natural gas system fuel use (i.e., lease, plant, and pipeline and distribution consumption) is assumed not to have additional end-use leakage not accounted for as a system leakage.

[3]Pipeline use is not differentiated between the transmission and distribution system. Given common uses of natural gas for compressor stations along the pipeline route, natural gas used for these purposes will be subject to some leakage from the systems they support (i.e., transmission or distribution) as well as upstream emissions.

[4]Approximate; exact relationships between users and the natural gas system are not clearly known. For example, some industrial users might be located on distribution lines.

combustion) or unintentional emissions in more dispersed settings (e.g., methane emissions from abandoned coal mines or nitrous oxide emissions from fertilized soils).

3.2. The location of methane leakage

Natural gas has unusually diverse consuming sectors: in the US, 40% is used for electricity generation and 30% each for industrial and residential/commercial uses (EIA, 2017). For LCA, this matters not only because it means that natural gas system methane leakage has pervasive GHG footprint implications (see Section 3.1.2), but also because not all parts of the natural gas supply system are used to serve all end users. Correct representations of a product system include methane leakage only from those upstream processes that are invoked by the product system. Studies of methane leakage often do not explicitly clarify which processes are appropriate for which types of end use, so this section provides rules of thumb for modeling the natural gas system.

Some processes are common to essentially all uses of natural gas. These include extraction, processing, and transmission through high pressure pipelines. Emissions from these processes can vary (e.g., by basin of origin, processing plant, or pipeline), but all natural gas users connected to the same infrastructure will induce the same upstream impacts.

Other processes are not common to all uses. The most significant divergence is that some end uses are served by low-pressure distribution systems downstream of high-pressure transmission systems, while others draw on the high-pressure system directly. Although both the transmission and distribution systems are sometimes called “pipelines” or “transportation,” they are separate systems. Thus, distribution leakage should not be allocated to users supplied directly by the transmission system. A rule of thumb is that natural gas used for power plants, industrial heat, and chemical plant feedstocks generally does not pass through the distribution system, while natural gas used for commercial and residential purposes generally does (Fig. 2, and see SM, S2). The most decision-relevant application of this observation is for natural gas-fired power plants, which are rarely fed by distribution lines, and which have frequently been compared to coal-fired power on the basis of relative carbon emissions. Given methane's high GWP, incorrectly including distribution leakage for natural gas-fired power can lead to nonneglible overestimates of GHG intensity (Alvarez et al., 2012).

We recommend that authors explicitly state which natural gas system processes are included in analysis, rather than citing an overall leakage rate. For high resolution studies, note that actual (rather than system-average) leakage can be highly site specific given different use of technologies, levels of training, safety and operational practices, year and/or season, legal context, company practice, or geology and land characteristics. Customer-side leakage is not well characterized by the empirical literature.

3.3. The influence of global warming potential on methane leakage impact

The scientific community's estimate of methane's GWP has not yet stabilized. Notably, published estimates have tended to increase over time, both for 20- and 100-year estimates (Fig. 3). For methane, IPCC-estimated GWP values that include climate-carbon feedback are slightly higher than GWP values that do not include this feedback, which is intended to account for inconsistent estimates of GWP for the reference gas (CO_2) versus other species (Gasser et al., 2017). The current IPCC estimate for methane's 100-year GWP (with climate-carbon feedback) is about 60% higher than the 1996 estimate and about 35% higher than the 2007 estimate; for the 20-year GWP, the current estimate is about 50% higher than the 1996 estimate and 20% higher than the 2007 estimate. The anticipated release of the Sixth Assessment Report in 2022—nearly ten years after the release of the current Fifth Assessment Report (AR5)—might update GWP estimates again.

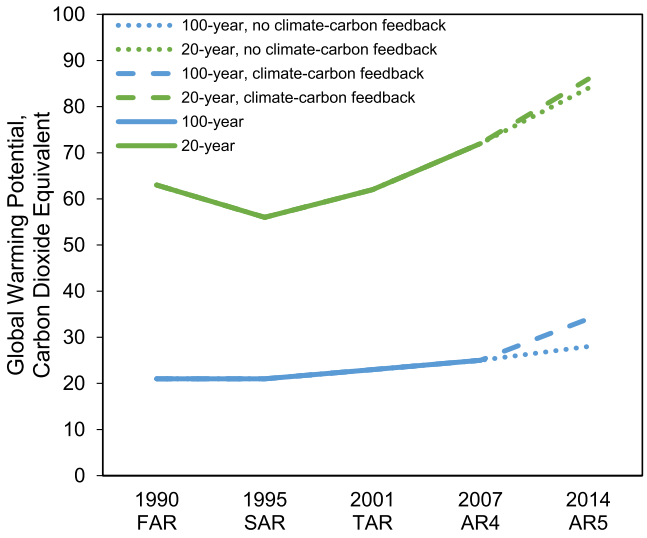

Fig. 3. Estimated global warming potential of methane by IPCC assessment report. **Data source:** Intergovernmental Panel on Climate Change (2018).

How much does GWP matter? Using a GWP of 25 (100-year, AR4), the 2015 GHGI estimates that natural gas systems accounted for 2.8% of total US GHG emissions (EPA, 2017). Simply updating the GWP to 34 (100-year, AR5 with climate-carbon feedback) raises this estimate to 3.6% of total US GHG emissions. Updating both GWP and natural gas system leakage rates to reflect state of the science estimates (Alvarez et al., 2018) suggests that methane leakage from natural gas systems alone accounts for 7.1% of total US GHG emissions—2.5 times the recorded estimate (EPA, 2017). Researchers using CO_2e-based literature estimates for methane impacts should take care to identify and harmonize GWP assumptions from previously published work. Especially for studies that directly address natural gas systems, conducting GWP sensitivity analysis is advisable.

4. Conclusion

The impact of methane leakage from natural gas systems is systematically underestimated and imprecisely characterized, which affects GHG footprints across product systems. Underestimated natural gas system methane leakage and low GWPs are both significant. LCA databases often underestimate leakage through key life cycle stages even relative to recorded estimates, and the connection between database estimates and relevant data is frequently unclear. The small number of processes involved, and the large impact to product LCA uncertainty, makes natural gas system methane leakage a priority target for data quality assurance in inventory databases. Meaningful improvements can be made with attention to the issues of leakage rates, system boundaries, and GWP described in this work.

Acknowledgements

Neither author has a conflict of interest to declare. This research did not receive any specific grant from funding agencies in the

public, commercial, or not-for-profit sectors.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2019.03.096.

References

Abrahams, L.S., Samaras, C., Griffin, W.M., Matthews, H.S., 2015. Life cycle greenhouse gas emissions from U.S. Liquefied natural gas exports: implications for end uses. Environ. Sci. Technol. 49, 3237–3245. https://doi.org/10.1021/es505617p.

Allen, D.T., Pacsi, A.P., Sullivan, D.W., Zavala-Araiza, D., Harrison, M., Keen, K., Fraser, M.P., Daniel Hill, A., Sawyer, R.F., Seinfeld, J.H., 2014. Methane emissions from process equipment at natural gas production sites in the United States: pneumatic controllers. Environ. Sci. Technol. https://doi.org/10.1021/es5040156.

Allen, D.T., Torres, V.M., Thomas, J., Sullivan, D.W., Harrison, M., Hendler, A., Herndon, S.C., Kolb, C.E., Fraser, M.P., Hill, A.D., Lamb, B.K., Miskimins, J., Sawyer, R.F., Seinfeld, J.H., 2013. Measurements of methane emissions at natural gas production sites in the United States. Proc. Natl. Acad. Sci. 110, 17768–17773. https://doi.org/10.1073/pnas.1304880110.

Alvarez, R.A., Pacala, S.W., Winebrake, J.J., Chameides, W.L., Hamburg, S.P., 2012. Greater focus needed on methane leakage from natural gas infrastructure. Proc. Natl. Acad. Sci. 109, 6435–6440. https://doi.org/10.1073/pnas.1202407109.

Alvarez, R.A., Zavala-Araiza, D., Lyon, D.R., Allen, D.T., Barkley, Z.R., Brandt, A.R., Davis, K.J., Herndon, S.C., Jacob, D.J., Karion, A., Kort, E.A., Lamb, B.K., Lauvaux, T., Maasakkers, J.D., Marchese, A.J., Omara, M., Pacala, S.W., Peischl, J., Robinson, A.L., Shepson, P.B., Sweeney, C., Townsend-Small, A., Wofsy, S.C., Hamburg, S.P., 2018. Assessment of methane emissions from the U.S. oil and gas supply chain. Science eaar7204. https://doi.org/10.1126/science.aar7204.

Balcombe, P., Brandon, N.P., Hawkes, A.D., 2018. Characterising the distribution of methane and carbon dioxide emissions from the natural gas supply chain. J. Clean. Prod. 172. https://doi.org/10.1016/j.jclepro.2017.11.223, 2019–2032.

Bouman, E.A., Ramirez, A., Hertwich, E.G., 2015. Multiregional environmental comparison of fossil fuel power generation—assessment of the contribution of fugitive emissions from conventional and unconventional fossil resources. Int. J. Greenh. Gas Control 33, 1–9. https://doi.org/10.1016/j.ijggc.2014.11.015.

Brandt, A.R., Heath, G.A., Cooley, D., 2016. Methane leaks from natural gas systems follow extreme distributions. Environ. Sci. Technol. 50, 12512–12520. https://doi.org/10.1021/acs.est.6b04303.

Brandt, A.R., Heath, G.A., Kort, E.A., O'Sullivan, F., Pétron, G., Jordaan, S.M., Tans, P., Wilcox, J., Gopstein, A.M., Arent, D., others, 2014. Methane leaks from North American natural gas systems. Science 343, 733–735.

Burnham, A., 2017. Updated Natural Gas Pathways in the GREET1_2017 Model. Argonne National Laboratory.

Burnham, A., Han, J., Clark, C.E., Wang, M., Dunn, J.B., Palou-Rivera, I., 2012. Life-cycle greenhouse gas emissions of shale gas, natural gas, coal, and petroleum. Environ. Sci. Technol. 46, 619–627. https://doi.org/10.1021/es201942m.

Carnegie Mellon University Green Design Institute, 2008. Economic Input-Output Life Cycle Assessment (EIO-LCA), US 2002 Producer Price Model [WWW Document]. URL. http://www.eiolca.net/index.html. (Accessed 18 January 2018).

Chang, Y., Huang, R., Ries, R.J., Masanet, E., 2015. Life-cycle comparison of greenhouse gas emissions and water consumption for coal and shale gas fired power generation in China. Energy 86, 335–343. https://doi.org/10.1016/j.energy.2015.04.034.

Costello, K.W., 2014. Lost and unaccounted-for gas: challenges for public utility regulators. Util. Pol. 29, 17–24. https://doi.org/10.1016/j.jup.2014.04.003.

ecoinvent, 2018. Ecoinvent Version 3.5 [WWW Document]. URL. https://www.ecoinvent.org/database/ecoinvent-35/ecoinvent-35.html. (Accessed 24 January 2019).

EIA, 2018. Table 1.3, Primary Energy Consumption by Source, Annual Energy Review.

EIA, 2017. U.S. Natural Gas Consumption by End Use [WWW Document]. URL. https://www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_a.htm. (Accessed 18 January 2018).

EPA, 2017. Inventory of U.S. Greenhouse Gas Emissions and Sinks, 1990-2015 (No. EPA 430-P-17-001).

EPA, 2015. Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2013 (Reports and Assessments).

Farquharson, D., Jaramillo, P., Schivley, G., Klima, K., Carlson, D., Samaras, C., 2017. Beyond global warming potential: a comparative application of climate impact metrics for the life cycle assessment of coal and natural gas based electricity. J. Ind. Ecol. 21, 857–873. https://doi.org/10.1111/jiec.12475.

Gallagher, M.E., Down, A., Ackley, R.C., Zhao, K., Phillips, N., Jackson, R.B., 2015. Natural gas pipeline replacement programs reduce methane leaks and improve consumer safety. Environ. Sci. Technol. Lett. 2, 286–291. https://doi.org/10.1021/acs.estlett.5b00213.

Gasser, T., Peters, G.P., Fuglestvedt, J.S., Collins, W.J., Shindell, D.T., Ciais, P., 2017. Accounting for the climate-carbon feedback in emission metrics. Earth Syst. Dyn. Discuss. 1–29. https://doi.org/10.5194/esd-2016-55.

Ge, X., Ma, J., Chang, L., Yuan, J., Su, X., Wang, H., Xiao, Y., 2016. Monitoring and investigating methane leakage in coal gas production. Pol. J. Environ. Stud. 25, 1005–1014. https://doi.org/10.15244/pjoes/61743.

Gibon, T., Arvesen, A., Hertwich, E.G., 2017. Life cycle assessment demonstrates environmental co-benefits and trade-offs of low-carbon electricity supply options. Renew. Sustain. Energy Rev. 76, 1283–1290. https://doi.org/10.1016/j.rser.2017.03.078.

Gilbert, A.Q., Sovacool, B.K., 2017. Benchmarking natural gas and coal-fired electricity generation in the United States. Energy 134, 622–628. https://doi.org/10.1016/j.energy.2017.05.194.

Gladd, K.H., 2016. New methane regulations for the oil and gas sector. Nat. Resour. Environ. Chic. 30, 51–53.

Grubert, E., 2017. Implicit prioritization in life cycle assessment: text mining and detecting metapatterns in the literature. Int. J. Life Cycle Assess. 22, 148–158. https://doi.org/10.1007/s11367-016-1153-2.

Hausfather, Z., 2015. Bounding the climate viability of natural gas as a bridge fuel to displace coal. Energy Policy 86, 286–294. https://doi.org/10.1016/j.enpol.2015.07.012.

Hausfather, Z., 2014. Climate Impacts of Coal and Natural Gas. Berkeley Earth.

Heath, G.A., O'Donoughue, P., Arent, D.J., Bazilian, M., 2014. Harmonization of initial estimates of shale gas life cycle greenhouse gas emissions for electric power generation. Proc. Natl. Acad. Sci. 111, E3167–E3176. https://doi.org/10.1073/pnas.1309334111.

Hendrick, M.F., Ackley, R., Sanaie-Movahed, B., Tang, X., Phillips, N.G., 2016. Fugitive methane emissions from leak-prone natural gas distribution infrastructure in urban environments. Environ. Pollut. 213, 710–716. https://doi.org/10.1016/j.envpol.2016.01.094.

Hopkins, F.M., Ehleringer, J.R., Bush, S.E., Duren, R.M., Miller, C.E., Lai, C.-T., Hsu, Y.-K., Carranza, V., Randerson, J.T., 2016. Mitigation of methane emissions in cities: how new measurements and partnerships can contribute to emissions reduction strategies. Earths Future 4, 408–425. https://doi.org/10.1002/2016EF000381.

Howarth, R.W., Santoro, R., Ingraffea, A., 2011. Methane and the greenhouse-gas footprint of natural gas from shale formations. A Lett. Clim. Change 106, 679–690. https://doi.org/10.1007/s10584-011-0061-5.

Intergovernmental Panel on Climate Change, 2018. Assessment Reports [WWW Document]. Intergov. Panel Clim. Change. URL. http://www.ipcc.ch/publications_and_data/publications_and_data_reports.shtml. (Accessed 18 January 2018).

Jackson, R.B., Down, A., Phillips, N.G., Ackley, R.C., Cook, C.W., Plata, D.L., Zhao, K., 2014. Natural gas pipeline leaks across Washington, DC. Environ. Sci. Technol. 48, 2051–2058. https://doi.org/10.1021/es404474x.

Jeong, S., Millstein, D., Fischer, M.L., 2014. Spatially explicit methane emissions from petroleum production and the natural gas system in California. Environ. Sci. Technol. 48, 5982–5990. https://doi.org/10.1021/es4046692.

Jiang, M., Griffin, W.M., Hendrickson, C., Jaramillo, P., VanBriesen, J., Venkatesh, A., 2011. Life cycle greenhouse gas emissions of Marcellus shale gas. Environ. Res. Lett. 6, 034014. https://doi.org/10.1088/1748-9326/6/3/034014.

Karion, A., Sweeney, C., Kort, E.A., Shepson, P.B., Brewer, A., Cambaliza, M., Conley, S.A., Davis, K., Deng, A., Hardesty, M., Herndon, S.C., Lauvaux, T., Lavoie, T., Lyon, D., Newberger, T., Pétron, G., Rella, C., Smith, M., Wolter, S., Yacovitch, T.I., Tans, P., 2015. Aircraft-based estimate of total methane emissions from the barnett shale region. Environ. Sci. Technol. 49, 8124–8131. https://doi.org/10.1021/acs.est.5b00217.

Karion, A., Sweeney, C., Pétron, G., Frost, G., Michael Hardesty, R., Kofler, J., Miller, B.R., Newberger, T., Wolter, S., Banta, R., Brewer, A., Dlugokencky, E., Lang, P., Montzka, S.A., Schnell, R., Tans, P., Trainer, M., Zamora, R., Conley, S., 2013. Methane emissions estimate from airborne measurements over a western United States natural gas field. Geophys. Res. Lett. 40, 4393–4397. https://doi.org/10.1002/grl.50811.

Kirchgessner, D.A., Lott, R.A., Michael Cowgill, R., Harrison, M.R., Shires, T.M., 1997. Estimate of methane emissions from the U.S. natural gas industry. Chemosphere 35, 1365–1390. https://doi.org/10.1016/S0045-6535(97)00236-1.

Lamb, B.K., Edburg, S.L., Ferrara, T.W., Howard, T., Harrison, M.R., Kolb, C.E., Townsend-Small, A., Dyck, W., Possolo, A., Whetstone, J.R., 2015. Direct measurements show decreasing methane emissions from natural gas local distribution systems in the United States. Environ. Sci. Technol. 49, 5161–5169. https://doi.org/10.1021/es505116p.

Lavoie, T.N., Shepson, P.B., Gore, C.A., Stirm, B.H., Kaeser, R., Wulle, B., Lyon, D., Rudek, J., 2017. Assessing the methane emissions from natural gas-fired power plants and oil refineries. Environ. Sci. Technol. 51, 3373–3381. https://doi.org/10.1021/acs.est.6b05531.

Lelieveld, J., Crutzen, P.J., Brühl, C., 1993. Climate effects of atmospheric methane. Chemosphere. In: Proceedings of the NATO advanced research workshop, 26, pp. 739–768. https://doi.org/10.1016/0045-6535(93)90458-H.

Lelieveld, J., Lechtenböhmer, S., Assonov, S.S., Brenninkmeijer, C.A.M., Dienst, C., Fischedick, M., Hanke, T., 2005. Greenhouse gases: low methane leakage from gas pipelines. Nature 434, 841–842. https://doi.org/10.1038/434841a.

Littlefield, J.A., Marriott, J., Schivley, G.A., Cooney, G., Skone, T.J., 2016. Using common boundaries to assess methane emissions: a life cycle evaluation of natural gas and coal power systems. J. Ind. Ecol. n/a-n/a https://doi.org/10.1111/jiec.12394.

McKain, K., Down, A., Raciti, S.M., Budney, J., Hutyra, L.R., Floerchinger, C., Herndon, S.C., Nehrkorn, T., Zahniser, M.S., Jackson, R.B., Phillips, N., Wofsy, S.C., 2015. Methane emissions from natural gas infrastructure and use in the urban

region of Boston, Massachusetts. Proc. Natl. Acad. Sci. 201416261. https://doi.org/10.1073/pnas.1416261112.

Meier, P.J., Wilson, P.P.H., Kulcinski, G.L., Denholm, P.L., 2005. US electric industry response to carbon constraint: a life-cycle assessment of supply side alternatives. Energy Policy 33, 1099–1108. https://doi.org/10.1016/j.enpol.2003.11.009.

Mitchell, A.L., Tkacik, D.S., Roscioli, J.R., Herndon, S.C., Yacovitch, T.I., Martinez, D.M., Vaughn, T.L., Williams, L.L., Sullivan, M.R., Floerchinger, C., Omara, M., Subramanian, R., Zimmerle, D., Marchese, A.J., Robinson, A.L., 2015. Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement results. Environ. Sci. Technol. 49, 3219–3227. https://doi.org/10.1021/es5052809.

NREL, 2012. U.S. Life Cycle Inventory Database [WWW Document]. URL. https://www.nrel.gov/lci/. (Accessed 18 January 2018).

Oliphant, M., 1994. Impact of Gas distribution leakage on gas appliance greenhouse effect. Clean Air J. Clean Air Soc. Aust. N. Z. 28, 24.

Pétron, G., Karion, A., Sweeney, C., Miller, B.R., Montzka, S.A., Frost, G.J., Trainer, M., Tans, P., Andrews, A., Kofler, J., Helmig, D., Guenther, D., Dlugokencky, E., Lang, P., Newberger, T., Wolter, S., Hall, B., Novelli, P., Brewer, A., Conley, S., Hardesty, M., Banta, R., White, A., Noone, D., Wolfe, D., Schnell, R., 2014. A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin. J. Geophys. Res. Atmos. 119, 2013JD021272. https://doi.org/10.1002/2013JD021272.

Phillips, N.G., Ackley, R., Crosson, E.R., Down, A., Hutyra, L.R., Brondfield, M., Karr, J.D., Zhao, K., Jackson, R.B., 2013. Mapping urban pipeline leaks: methane leaks across Boston. Environ. Pollut. 173, 1–4. https://doi.org/10.1016/j.envpol.2012.11.003.

Ravikumar, A.P., Brandt, A.R., 2017. Designing better methane mitigation policies: the challenge of distributed small sources in the natural gas sector. Environ. Res. Lett. 12, 044023. https://doi.org/10.1088/1748-9326/aa6791.

Roy, M., Edwards, M.R., Trancik, J.E., 2015. Methane mitigation timelines to inform energy technology evaluation. Environ. Res. Lett. 10, 114024. https://doi.org/10.1088/1748-9326/10/11/114024.

Sanchez, N., Mays, D.C., 2015. Effect of methane leakage on the greenhouse gas footprint of electricity generation. Clim. Change 133, 169–178. https://doi.org/10.1007/s10584-015-1471-6.

von Fischer, J.C., Cooley, D., Chamberlain, S., Gaylord, A., Griebenow, C.J., Hamburg, S.P., Salo, J., Schumacher, R., Theobald, D., Ham, J., 2017. Rapid, vehicle-based identification of location and magnitude of urban natural gas pipeline leaks. Environ. Sci. Technol. 51, 4091–4099. https://doi.org/10.1021/acs.est.6b06095.

Weyant, J.P., de la Chesnaye, F.C., Blanford, G.J., 2006. Overview of EMF-21: multigas mitigation and climate policy. Energy J. 27, 1–32.

Yacovitch, T.I., Neininger, B., Herndon, S.C., Van der Gon, H.D., Jonkers, S., Hulskotte, J., Roscioli, J.R., Zavala-Araiza, D., 2018. Methane emissions in The Netherlands: the Groningen field. Elem Sci Anth 6, 57. https://doi.org/10.1525/elementa.308.

Zhang, X., Myhrvold, N.P., Caldeira, K., 2014. Key factors for assessing climate benefits of natural gas versus coal electricity generation. Environ. Res. Lett. 9, 114022. https://doi.org/10.1088/1748-9326/9/11/114022.

Zimmerle, D.J., Williams, L.L., Vaughn, T.L., Quinn, C., Subramanian, R., Duggan, G.P., Willson, B., Opsomer, J.D., Marchese, A.J., Martinez, D.M., Robinson, A.L., 2015. Methane emissions from the natural gas transmission and storage system in the United States. Environ. Sci. Technol. 49, 9374–9383. https://doi.org/10.1021/acs.est.5b01669.